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IGNITION OF ALUMINUM PARTICLES AND CLOUDS

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Abstract

Here we review experimental data and models of the ignition of aluminum (Al) particles and clouds in explosion fields. The review considers: (i) ignition temperatures measured for single Al particles in torch experiments; (ii) thermal explosion models of the ignition of single Al particles; and (iii) the unsteady ignition Al particles clouds in reflected shock environments. These are used to develop an empirical ignition model appropriate for numerical simulations of Al particle combustion in shock dispersed fuel explosions.

1. Introduction

We consider the problem of combustion of aluminum (Al) particle clouds in explosions from Shock-Dispersed-Fuel (SDF) charges [1]. The charge consists of a spherical PETN booster (1/3 the charge mass), surrounded by aluminum powder (2/3 the charge mass) at an initial density of 0.6 g/cc. Detonation of the booster charge creates a blast wave that disperses the Al powder and ignites the ensuing Al-air mixture—thereby forming a two-phase combustion cloud embedded in the explosion. Afterburning of the booster detonation products with air also enhances and promotes the Al-air combustion process. Pressure waves from such reactive blast waves have been measured in bomb calorimeter experiments [1, 2]. The dynamics of energy release in Al-SDF explosions has been simulated with our heterogeneous continuum combustion model [3]. Such simulations require a model of aluminum particle ignition and combustion as an input. Here we provide a literature review of aluminum particle ignition in support of such modeling. It is based on:

- Torch experiments on the ignition of single Al particles by Gurevich et al [3]
- Thermal explosion model of Al particle ignition by Federov & Kharlamova [4]
- Ignition of Al particle clouds in reflected shock environments by Boiko et al [5,6]

These will be used to construct an empirical model of the ignition of aluminum particle clouds.

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2. Ignition of Single Aluminum Particles

Ignition limits of aluminum particles have been studied in powder-burning torch experiments by Gurevich, Lapinka and Ozerov [3]. They measured the ignition temperature of aluminum particles in oxygen diluted with argon and nitrogen. Results are summarized in Fig. 1 which shows that particle ignition temperature, T_{ign} , depends on the particle diameter, d , and oxygen concentration, C_{O_2} . Measured ignition temperatures ranged from 2,057 C (consistent with the melt temperature of Al_2O_3) down to 696 C (near the melt temperature of pure aluminum: 660 C). Data points were fit with the following function [3]:

$$T_{ign} = f_G(d, C_{O_2}) = T_{mpo} - 0.6 - \frac{C_{O_2}^{0.3} d}{\lambda} \exp(-0.85\sqrt{d}) \quad (1)$$

where T_{mpo} is the melting point of the aluminum oxide coating ($\sim 2,300$ K) and λ is the thermal conductivity of the gas. The minimum measured ignition temperature was 696 C for $d = 6\mu m$ particles near stoichiometric conditions.

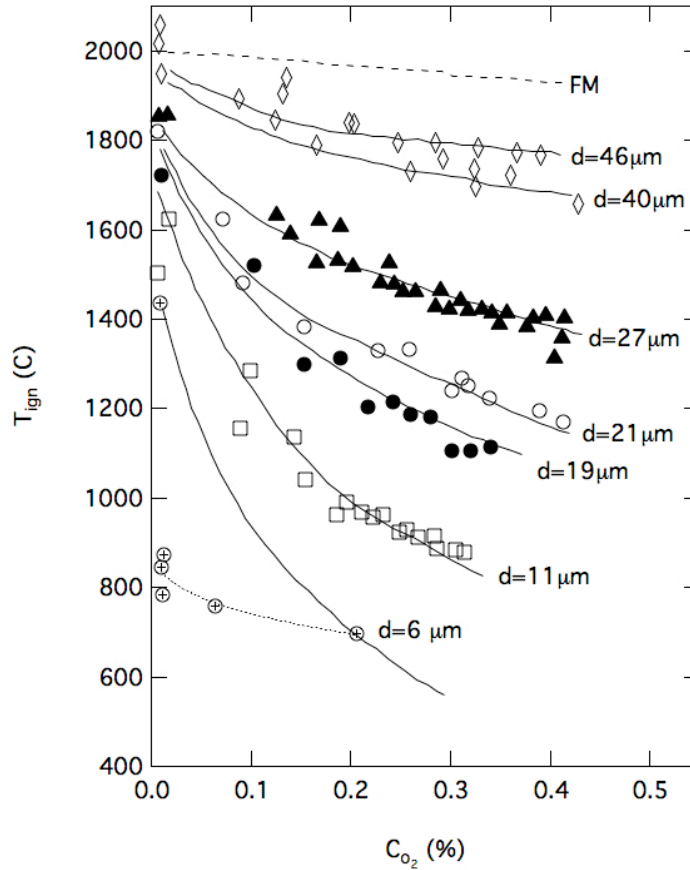


Figure 1. Ignition temperature for a single aluminum particle in oxygen diluted with argon and nitrogen, as measured by Gurevich et al (1968). Curves represent Eq. (1) of Gurevich; dashed line FM denotes results of Friedman and Maček (1962); dotted line is an alternate fit to the $d = 6\mu m$ data points.

3. Thermal Explosion Model

Merzhanov [8] has laid down the fundamentals of a thermal theory of metal particle ignition. Here we follow the work of Federov and Kharlamova [4] who have applied the theory to model the ignition of a single aluminum particle in oxygen. Results are presented in Fig. 2, which shows the ignition delay for a $6\mu\text{m}$ particle at different atmospheric temperatures.

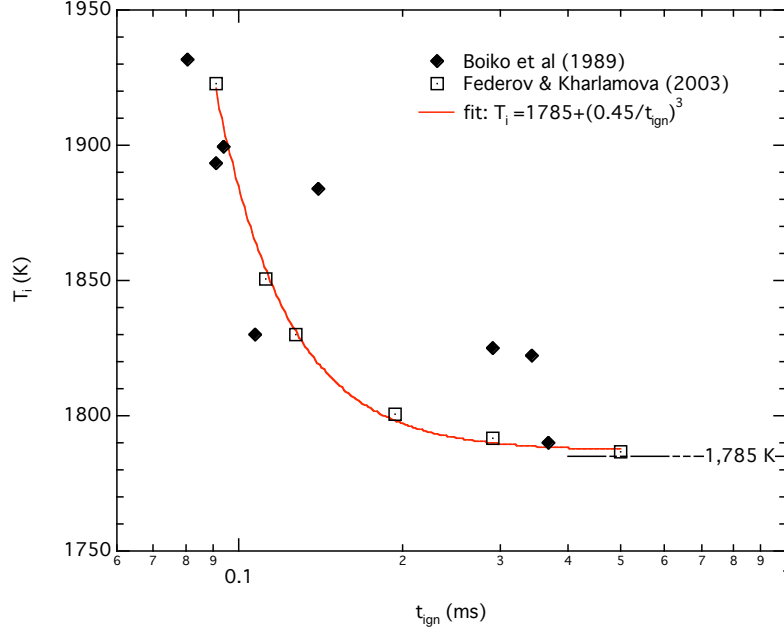


Figure 2. Ignition delay t_{ign} at different atmospheric temperatures for a $6\mu\text{m}$ spherical Al particle in oxygen, according to the thermal explosion model of Federov & Kharlamova (2003).

Results are consistent with some of the data points from the experiments by Boiko et al [5], [6]. The model predicts a minimum ignition temperature of 1,785K for a $6\mu\text{m}$ particle. This value is consistent with one Gurevich data point (1,710K) for very low oxygen concentrations ($C_{O_2} = 0.009\%$). Model values of ignition temperatures (square symbols) have been fit with the following function:

$$T_i(K) = 1,785 + (0.45/t_{ign})^3 \quad (2)$$

This may be inverted to express the ignition delay as a cube-root function of temperature:

$$t_{ign}(ms) = 0.45/(T_i - 1,785)^{1/3} \quad \text{for } 1,785K < T_i < 2,800K \quad (3)$$

Ignition delays ranged from $45\mu\text{s}$ at 2,800K to infinity at 1,785K.

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Ignition delays for various particle diameters were also predicted by the thermal explosion model [4]; results are presented in Fig. 3. Predicted ignition delays are in agreement with the data of Pokhil et al [9]. Model results were fit with the following function

$$t_{ign}(ms) = 8.6 \times 10^{-4} d^{2.3} \quad (4)$$

with d in μm . It exhibits a 2.3 power dependence on particle diameter. For $6 \mu m$ particles, it predicts an ignition delay of $53 \mu s$.

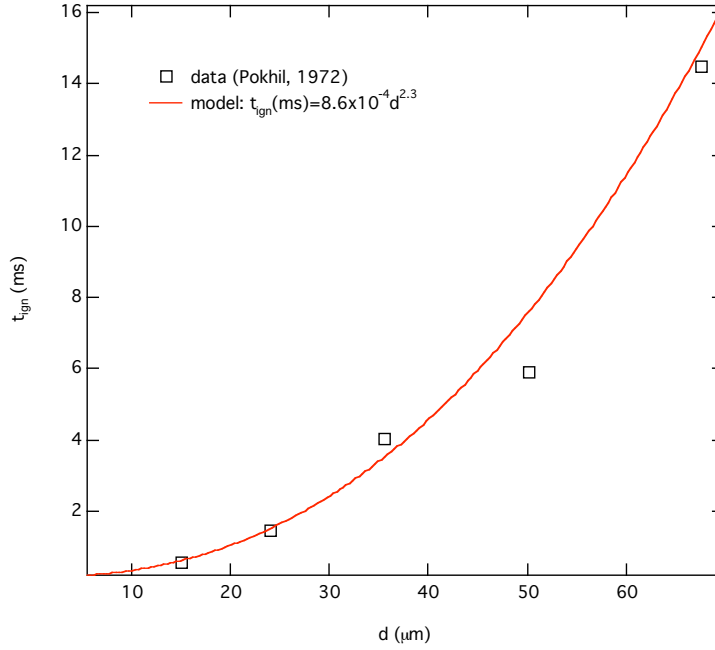


Figure 3. Ignition delay is plotted as a function of aluminum particle diameter; data are from Pokhil et al (1972), which are modeled by Federov & Kharlamova (2003) and fit with a power-law function (4).

4. Ignition of Aluminum Particle Clouds

Boiko and co-workers [5], [6] used streak photography to visualize the ignition of aluminum particle clouds in reflected shock environments in a shock tube. A typical example of aluminum cloud ignition at $T = 1900$ K and $p = 1.1$ MPa is presented in Fig. 4. One can see that the *cloud ignition* depends, not only on the pressure and temperature and oxygen concentration (as in single particle ignition), but also on the fuel mass loading of the cloud. For low fuel loadings (Fig. 4c), individual particles can ignite but they burn out without igniting nearby particles. At higher fuel loadings, their concentration is large enough to ignite the entire cloud (Figs. 4a,b); Boiko calls this the *self-ignition regime* of aluminum cloud combustion. This figure also shows that the ignition delay is independent of the fuel loading (i.e., ignition delay is the same in Figs. 4a, 4b and 4c).

Ignition of aluminum particle clouds in the low-temperature regime was also observed experimentally [5, 6]; this was labeled the *abnormal ignition regime* (because it does not agree with steady-state single-particle data). An example of ignition in this low-temperature regime ($T = 1370$ K) is shown in Fig. 5. For a mass loading of 7 mg (Fig. 5a), one can see a few particles ignite but then burn out. For a mass loading of 10 mg (Fig. 5b), it appears that a single particle ignites the entire cloud at 3 ms.

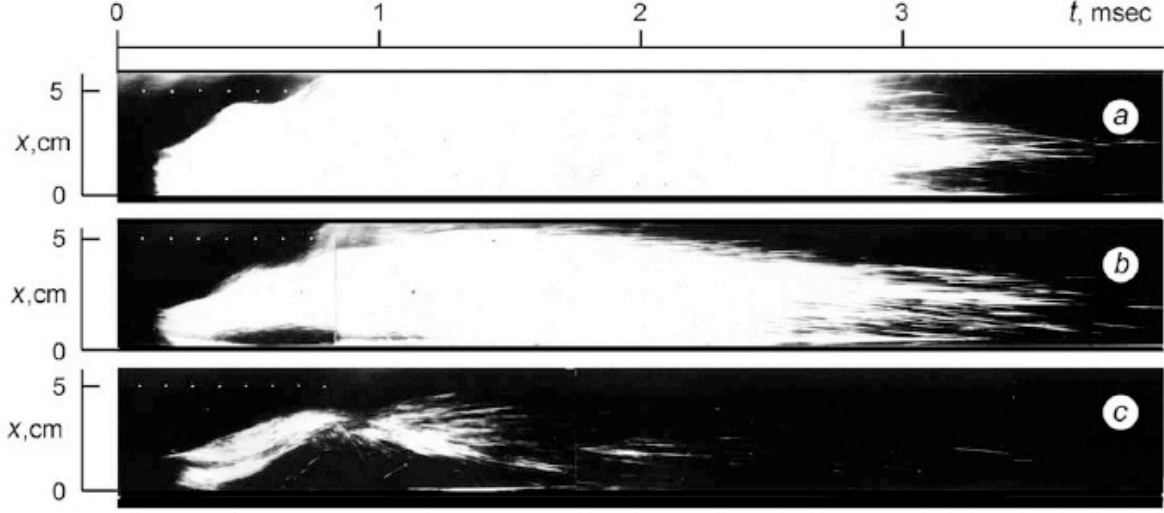


Figure 4. Streak photography of the ignition of an aluminum particle cloud at $T = 1900$ K and $p = 1.1$ MPa for different fuel loading, m : (a) $m = 5$ mg, (b) $m = 1$ mg, (c) $m = 0.25$ mg (Boiko & Poplavski, 2002).

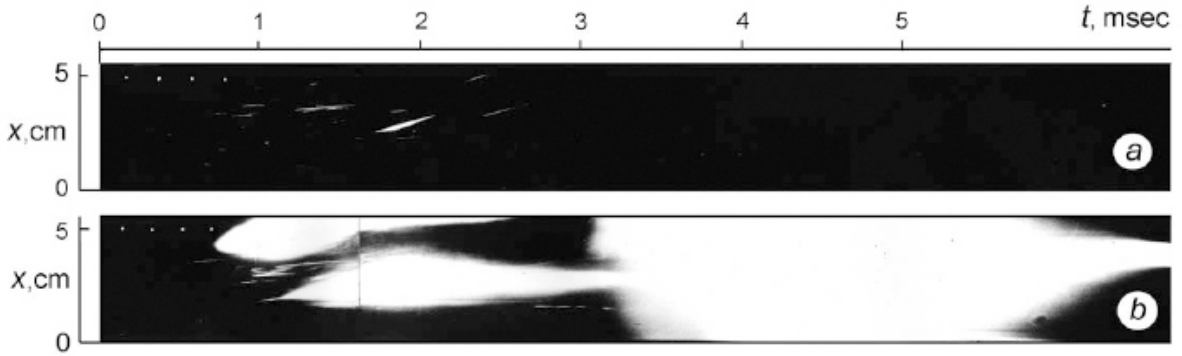


Figure 5. Streak photography of the ignition of an aluminum particle cloud at $T = 1370$ K and $p = 2.5$ MPa for different fuel mass loadings, m : (a) $m = 7$ mg, (b) $m = 10$ mg (Boiko & Poplavski, 2002).

Ignition delays were measured from the streak photography; results are presented in Fig. 6. Data in the self-ignition regime (open symbols) correlate with the Friedman-Mačec model of particle ignition in Bunsen-burner flames [7], namely:

$$\tau_* = f_{FM}(T, d) = \frac{\rho_s d^2}{12\lambda(T)} \left(c_s \ln \frac{T - 290}{T - T_*} + \frac{\Delta H}{T - T_m} \right) \quad (5)$$

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where $\rho_s = 2.7 \text{ g/cc}$ for Al, $c_s = 1 \text{ J/g} \cdot \text{K}$ is the gas specific heat capacity, $T_m = 933 \text{ K}$ is the melt temperature of Al, $\Delta H = 400 \text{ J/g}$ is the heat of fusion for pure Al, $T =$ reflected-shock gas temperature, $T_* =$ Al-particle ignition temperature, $\lambda(T) = \lambda_0(T/290)^{3/4} =$ thermal conductivity of the gas with $\lambda_0 = 2.4 \cdot 10^{-2} \text{ J/(m} \cdot \text{s} \cdot \text{K)}$. Particles in the self-ignition regime (open symbols) correlate with an Al-particle ignition temperature of $T_* = 1,800 \text{ K}$ (solid curve in Fig. 6).

Ignition of Al particles at temperatures considerably below 1,800 K—even as low as 1,000 K—was also observed (closed symbols of Fig. 6). The latter points agree with the Guervich measurements (993K—1,133K) for $d = 6\mu\text{m}$ aluminum particles (dotted curve of Fig. 1). In these cases, only a small fraction of the Al particles in the cloud ignited. Boiko ascribed this to uncontrolled properties of the particles (e.g., cracks, variation in the Al_2O_3 oxide coating thickness, number of ultra fine particles, etc.) in the test sample taken from same batch of Al powder. Apparently, a high Al-particle concentration is one of the conditions for ignition in the low temperature regime [6].

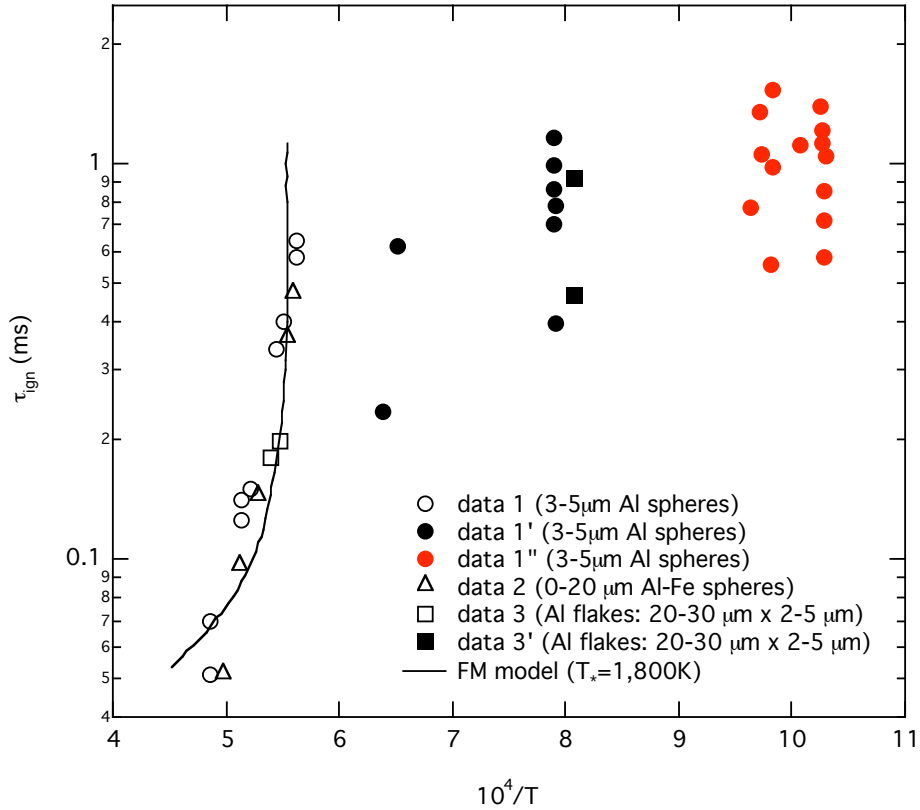


Figure 6. Ignition delay for aluminum particle clouds as measured by Boiko et al (1989); particle group 1: $d = 3 - 5\mu\text{m}$ Al spheres; group 2: $d = 0 - 20\mu\text{m}$ spherical particles of Al (89%) and Fe (11%); group 3: Al flakes, size = $20 - 30\mu\text{m}$ and thickness = $1 - 5\mu\text{m}$; solid curve = eq. (5).

For consistency, we analyze the ignition delay data in the self-ignition regime ($\sim 1,800$ K) of Fig. 6; data points are presented in an *Arrhenius-type* plot in Fig. 7.

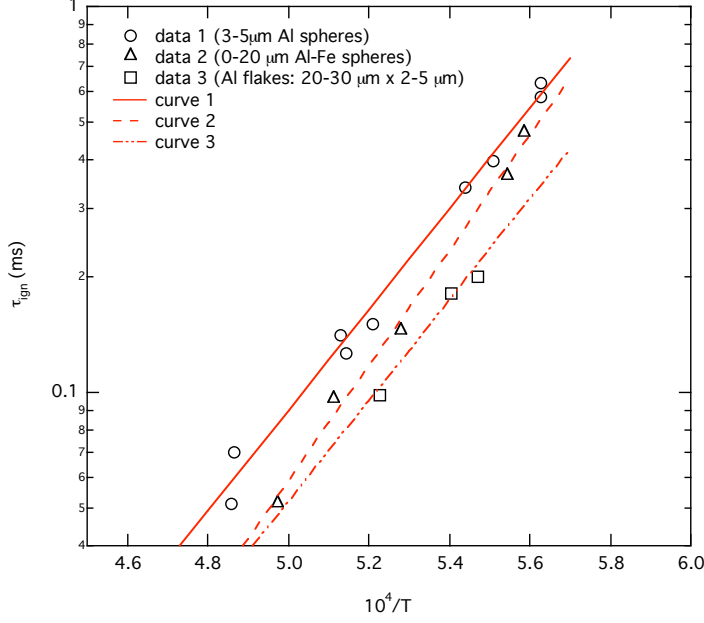


Figure 7. Measured ignition delays from Fig. 5 in the “normal ignition” regime ($T_* \sim 1,800$ K) curve 1: $\tau_1 = 2.5 \times 10^{-8} e^{30,000/T}$, curve 2: $\tau_2 = 0.2 \times 10^{-8} e^{34,400/T}$; curve 3: $\tau_3 = 1.6 \times 10^{-8} e^{30,000/T}$.

Results were fit with exponential functions of temperature:

$$\text{spherical Al particles:} \quad \tau_1 = 2.5 \times 10^{-8} e^{30,000/T} = 2.5 \times 10^{-8} e^{60,000/RT} \quad (6)$$

$$\text{flake Al particles:} \quad \tau_3 = 1.6 \times 10^{-8} e^{30,000/T} = 1.6 \times 10^{-8} e^{60,000/RT} \quad (7)$$

The above correlations imply a global activation-energy of $E_a = 60$ kCal/mole for Al particles. Note that ignition delays for flake Al particles are 60% shorter than ignition delays for spherical Al particles—a geometric effect on ignition. Coming from data in the self-ignition regime (open symbols in Fig. 6), one could say that these fits are also consistent with the Friedman-Maček function: $f_{FM}(T, d)$.

5. Cloud Ignition Model

The shock tube experiments [5, 6] prove conclusively that the ignition of aluminum particle clouds depends on the fuel concentration. If the fuel loading density is too small, a few particles may ignite but they burn out without igniting the whole cloud (Figs. 4c and Fig. 5a). One can formulate this effect as a probability of cloud ignition, $\mu_c(\rho_F)$. Go/no-go data from Fig. 4 were used to construct the cloud ignition probability as a function of the fuel density, ρ_F , based on an estimated cloud volume of 5 cm^3 . Results are presented in Fig. 8.

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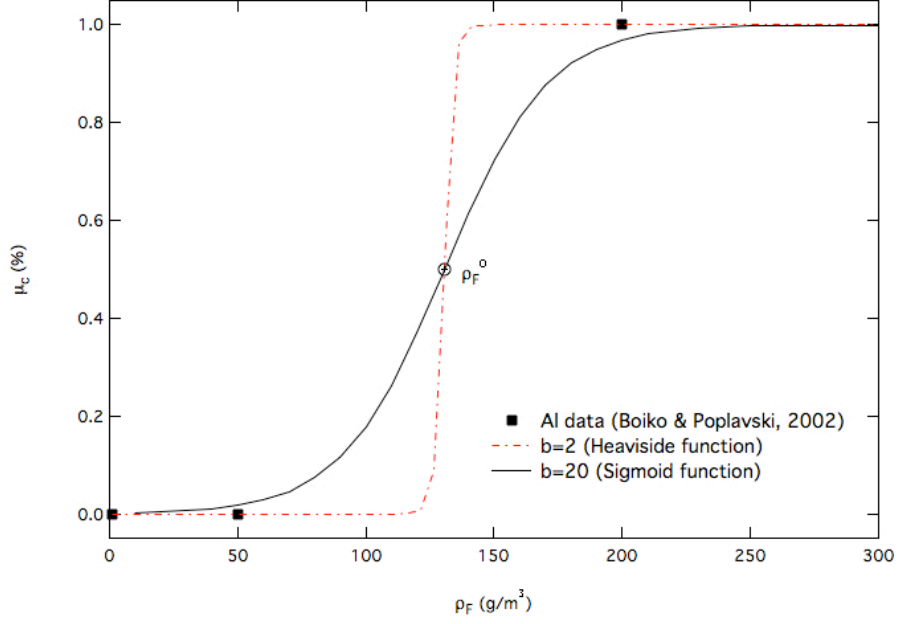


Figure 8. Ignition probability for a cloud of aluminum particles in the self-ignition regime ($T_* \sim 1,800K$), as a function of fuel loading: ρ_F , based on the shock tube experiments [6].

The data were used to construct the cloud ignition probability function:

$$\mu_c(\rho_F) = \frac{1}{1 + \exp[(\rho_F^0 - \rho_F)/b]} \quad (8)$$

where $[\rho_F] = g/m^3$. The function contains two fitting constants: ρ_F^0 and b . The first corresponds to the critical fuel density for 50% probability of cloud ignition (in the current case, $\rho_F^0 = 130 g/m^3$). The second corresponds to the width parameter, b . For small values of b , the probability function approaches a Heaviside function ($b = 2$ in Fig. 8); for larger values ($b = 20$) the probability function is a Sigmoid function in Fig. 8.

6. Cloud Combustion Model

Measured ignition delays (6)-(7) and ignition probabilities (8) can be used as a basis of constructing an empirical ignition model for aluminum particle clouds. Following Koribeinikov et al [10] and Oran et al [11] for gas phase systems, let $f(\mathbf{x}, t)$ denote the fraction of ignition time elapsed at time t . For a Lagrangian particle, it satisfies the following kinetics equation:

$$\frac{df}{dt} = \frac{1}{\tau_{ign}(T)} = Ae^{-E_a/RT} \quad (9)$$

For flake aluminum particles, $A = 6.25 \times 10^7$ and $E_a = 60 \text{ k-Cal/mole}$, based on fit (7).

Re-stating this for a Eulerian field, one finds:

$$\partial_t f + \mathbf{u} \cdot \nabla f = A e^{-E_a/RT} \quad (10)$$

with an initial condition of $f(\mathbf{x}, 0) = 0$. One can then allow ignition in a computational cell at location \mathbf{x} at time t when $f(\mathbf{x}, t) \geq 1$. Once ignited (i.e., $f = 1$), Al-air combustion products are produced according to the mass conservation law:

$$\partial_t \rho Y_p + \nabla \cdot \rho Y_p \mathbf{u} = \mu_c(\rho_F) \cdot (1 + \alpha_s) \cdot \dot{s} \quad (f = 1) \quad (11)$$

where ρ is the gas density, Y_p is the mass-fraction of combustion products, \mathbf{u} is the particle velocity vector, α_s is the stoichiometric air/fuel ratio, and \dot{s} is the reaction rate. Here, the rate of combustion is controlled by the rate of oxygen is supplied by turbulent mixing. The probability of cloud ignition is taken into account by μ_c . The applicability of this model will be explored in future numerical simulations.

7. Summary

Bunsen burner experiments of single aluminum particles in oxygen show that the ignition temperature depends on the particle diameter and oxygen concentration: $T_{ign} = f_G(d, C_{O_2})$. For large particles ($d > 46 \mu m$) and low oxygen concentrations, measured ignition temperatures were around 2330 K, consistent with the melt temperature of aluminum oxide. For small particles ($d \leq 6 \mu m$) and low oxygen concentrations, ignition temperatures of 1053 K—1147 K were measured. Near stoichiometric conditions, an ignition temperature of 969 K was measured for $6 \mu m$ particles; this is close to the melt temperature of pure aluminum (933 K). Intermediate particle diameters and oxygen concentrations yield ignition temperatures between these two limits (969 K—2330 K), according to the Gurevich function $f_G(d, C_{O_2})$.

The thermal explosion model of Federov predicts a minimum ignition temperature of 1785 K for $6 \mu m$ particles in oxygen; this is consistent with the data point: 1710 K measured by Gurevich for $6 \mu m$ particles at $C_{O_2} = 0.009\%$. Ignition delay increased as the 2.3-power of the particle diameter {see eq. (4)}; for $6 \mu m$ particles, it predicts an ignition delay of $53 \mu s$.

Shock-induced ignition of aluminum particle clouds was measured in reflected shock environments [5, 6]. In the self-ignition regime, the ignition delay correlated with the Friedman-Maček function $f_{FM}(T, d)$. It gave a characteristic ignition temperature of $T_* = 1,800$ K, consistent with the Federov model (1,785K) and Gurevich measurement

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(1,710K) for $6\ \mu\text{m}$ particles. Ignition delay followed an Arrhenius dependence on temperature: $\tau_1(\text{ms}) = 2.5 \times 10^{-8} e^{60,000/RT}$ for $3-5\ \mu\text{m}$ spherical Al particles; this dependence implies an activation-energy of 60 kcal/mole for aluminum particles in oxygen. These experiments also proved that the ignition of an Al particle cloud depends on the fuel loading. We have modeled this effect by a cloud ignition probability function: $\mu_c(\rho_F) = [1 + \exp\{(\rho_F^0 - \rho_F)/b\}]^{-1}$. Although individual particles may ignite, the entire cloud will not ignite unless the fuel loading exceeds a critical value: ρ_F^0 . Based on limited data sets [5, 6], we estimate $\rho_F^0 = 130\ \text{g}/\text{m}^3$ for the Al particles in the self-ignition regime ($T_* \sim 1,800\text{K}$). More data (and analysis) is needed to establish the cloud ignition probability function in the low-temperature abnormal-ignition regime ($933\ \text{K} < T < 1800\ \text{K}$).

8. Conclusions

Ignition of aluminum particle clouds in SDF explosions involves an additional level of complexity due to the inherent non-steadiness of the problem. Particles are embedded in a blast wave driven by the detonation of the booster charge. Initially, they can be ignited by the hot detonation products from the booster, or by its hot combustion products with air. However, this heat source is subjected to strong cooling effects induced by the blast wave expansion. Thus particle heating is fundamentally unsteady. Viewed from the thermal explosion perspective, ignition becomes a temporal competition—a race—between the unsteady heating by the surrounding gases and the exothermic effects of the Al-air combustion, and unsteady cooling due to blast wave expansion and entrainment of cool air due to turbulent mixing. Such unsteady effects favor the ignition of small particles over large particles. The fate of the entire aluminum particle cloud then depends on a collective competition over all scales of the particle ensemble of the cloud, along with a fuel loading large enough to support sustained combustion.

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